

Dependence of Intake Fraction on Release Location in a Multi-media Framework: A Case Study of Four Contaminants in North America

*Matthew MacLeod^{φ,§}, Deborah H. Bennett[†], Merike Perem^φ,
Randy L. Maddalena[§], Thomas E. McKone^{§,Ψ} and Don Mackay^φ

^φCanadian Environmental Modelling Centre
Trent University
Peterborough, Ontario, Canada

[†]School of Public Health,
Harvard University
Boston, MA

[§]Environmental Energy Technologies Division
Lawrence Berkeley National Laboratory
Berkeley, CA

^Ψ School of Public Health
University of California, Berkeley
Berkeley, CA

March 2003

This work was supported by the Natural Sciences and Engineering Research Council of Canada (NSERC) and by the US Environmental Protection Agency National Exposure Research Laboratory through Interagency Agreement # DW-988-38190-01-0, and Kresge Center for Environmental Health, NIEHS, through the U.S. Department of Energy under Contract Grant No. DE-AC03-76SF00098.

Dependence of Intake Fraction on Release Location in a Multi-media Framework: A
Case Study of Four Contaminants in North America

*Matthew MacLeod^{φ,§}, Deborah H. Bennett[†], Merike Perem^φ, Randy L. Maddalena[§],
Thomas E. McKone^{§,Ψ} and Don Mackay^φ

(φ) Canadian Environmental Modelling Centre, Trent University

(†) School of Public Health, Harvard University

(§) Environmental Energy Technologies Division, Lawrence Berkeley National
Laboratory

(Ψ) School of Public Health, University of California, Berkeley

Abbreviated form of the title:

Variability in Intake Fraction

Corresponding Author:

*Matthew J. MacLeod

Lawrence Berkeley National Laboratory

One Cyclotron Road, 90R3058

Berkeley, CA

94720-8132

mjmacleod@lbl.gov

Phone: 510 486-6945

FAX: 510 486-6658

Keywords: Multimedia modeling, Life cycle analysis, Comparative impact assessment,
Exposure, Dietary exposure pathway

Abstract

The extent of human exposure to persistent anthropogenic environmental contaminants is a complex function of the amount of chemical emitted, its physico-chemical properties and reactivity, the nature of the environment, and the characteristics of the pathways for human exposure, such as inhalation, intake of food and water and dermal contact. For some chemicals, the location of emissions relative to areas of high population density or intense food production may also be an important factor. The relative importance of these variables is explored using the regionally segmented BETR North America contaminant fate model and data for food production patterns and population density for North America. The model is applied to four contaminants emitted to air: benzene, carbon tetrachloride, benzo[a]pyrene and 2,3,7,8-tetrachlorodibenzo dioxin. The total continental intake fraction (iF), relating exposure quantity to emission quantity, is employed as a metric for assessing population exposure to environmental contaminants. The results show that the use of continentally averaged parameters for population density and food production provides an accurate estimate of the median of iF calculated for emissions in individual regions, however iF can range from this median by up to 3 orders of magnitude, especially for chemicals transferred to humans through the food pathway. The location of population relative to food production and emissions of chemicals are important variables that should be considered in assessing the public health implications of chemical emissions.

INTRODUCTION

Given the range of possible pathways for human exposure and the large number of industrial, agricultural and consumer activities that release chemicals into the environment, simplified methods are needed to assess different scenarios and to identify the combinations of chemical, environmental and population characteristics that lead to elevated exposures. Models of the environmental fate of chemical contaminants and subsequent human exposure have therefore found a variety of scientific and regulatory applications. These include screening level environmental fate assessments, comparative risk assessment of chemicals, and life cycle impact assessments of product or process alternatives. Screening assessments often use intake of chemical as a convenient metric of potential dose of contaminant received by a population. The population-based intake of an environmental contaminant depends on 1) the receiving media for the emission and the multi-media fate and transport of the chemical in the environment, 2) the relationship between contaminant concentration in environmental media and exposure media and 3) the rate at which the population contacts the exposure media through multiple exposure pathways.

There is an incentive to improve current models that describe the source-to-intake relationship for pollutants, and to build confidence in the models by evaluating their performance against available monitoring data. With access to reliable data and models that have been endorsed through peer review, regulators evaluating new chemicals could identify those that pose significant hazard prior to their introduction into the environment. Predictive models also provide a framework for comparing the “toxicity potential” of industrial products and processes in life cycle impact assessments comparing two or more alternative processes and their associated chemical release scenarios.

Intake fraction (iF) has been proposed as an informative and simple metric for interpreting complex fate and exposure calculations by conveying the source-to-intake relationship for different emission scenarios (Bennett, et al. 2002b). The iF (dimensionless) under the assumption of steady-state conditions is defined as the ratio of

the rate of pollutant intake by the population (mg/day) to the rate of release into the environment (mg/day). It is the fraction of the quantity emitted that enters the human population. In principle iF could be calculated using emissions inventories and monitoring studies, however in practice significant data gaps require estimation of emission rates and the use of models to describe the environmental fate and human exposure to the pollutant.

Several generic screening-level models have been developed to identify chemical and exposure characteristics that result in elevated exposures (Brandes, et al. 1996, Huijbregts, et al. 2000a, McKone 1993). A common feature of these models is that they do not include a spatially resolved description of the environment and the population. They treat each environmental medium as homogeneous with respect to the chemical concentration within the region under consideration. Population density and intensity of food production are likewise assumed to be uniform within the region.

Neglecting spatial heterogeneity introduces a degree of model uncertainty into the assessment because certain characteristics of the real system are not captured. For example, if exposure to a contaminant is primarily through inhalation, iF will differ for a given pollutant release in a densely populated urban region compared to the same release in a sparsely populated region. Similarly, if exposure is primarily through ingestion, iF is likely to differ due to the proximity of the source to agricultural regions, where contaminants enter the dietary exposure pathway. The properties of the chemical and environmental factors such as atmospheric conditions or the presence or absence of water bodies in a region will also affect environmental fate and transfer of contaminants into human exposure pathways. Since these effects are potentially important and are not accounted for in a non-spatially explicit multimedia model, there is an incentive to develop models accounting for spatial differences in environmental characteristics, population, food production and contaminant concentrations.

In this paper we present a spatially explicit multi-media contaminant fate and human exposure model for assessing source-to-intake relationships in North America, and

illustrate it by application to four contaminants that span a range of environmental mobility and exposure characteristics. The model framework relies on regional-scale databases for North American food production activities that have been compiled from a survey of agricultural statistics. The spatially explicit model framework presented here is a refinement of existing models that describe the environment and population using a single set of average characteristics. We evaluate the importance of the location of pollutant sources relative to regions of agricultural production and high population density when conducting population-based exposure assessments using iF.

METHODS

We have developed a human exposure assessment module that can be coupled with the existing Berkeley-Trent North American contaminant fate model (BETR North America) (MacLeod, et al. 2001, Woodfine, et al. 2001). BETR North America describes the North American environment as 24 ecological regions, as illustrated in Figure 1. Within each region the fate of environmental contaminants is described using a seven-compartment fugacity model that calculates multi-media environmental distribution between two atmospheric layers, vegetation, soil, freshwater, freshwater sediments and coastal water. The model accounts for transport of contaminants between adjacent regions in the atmosphere, in rivers and in near-shore ocean currents. The contaminant fate model is based on the mass balance principle; the total amount of contaminant that enters each region and the continental environment as a whole is tracked and balanced with removal processes. The BETR North America contaminant fate model is applied here to deduce steady-state contaminant concentrations in the environment.

Adapting and extending the BETR North America contaminant fate model for human exposure calculations required formulating equations to describe transfer of contaminants from the environment into pathways for human exposure, development of a database of population and food production activities in North America, and interpretation of the results using iF. Methods used in each of these activities are described below.

BETR North America Human Exposure Calculations

The human exposure module for the BETR North America model is based on a multiple pathway exposure assessment that links chemical concentrations in environmental media calculated by the contaminant fate model to exposure concentrations and contact rates for individuals through inhalation, ingestion and dermal contact. Ingestion pathways include consumption of four classes of vegetation (grains, protected produce, leafy vegetables and root crops), five classes of animal products (beef and poultry, eggs, fish, and dairy products), potable water consumption and incidental soil ingestion. Dermal exposure is from contaminated water during bathing and recreation, and contact with soil. This pathway is only significant for a very limited number of pollutants.

Many equations and input parameters describing individual exposure pathways have been taken from the most recent version of the CalTOX model (CalTOX 2001), which has been widely and successfully applied in human exposure and risk assessments of environmental pollutants (Bennett, et al. 2002a, Bennett, et al. 1998, Hertwich, et al. 2001). However, exposure concentrations in the agricultural food chain are treated differently in the BETR exposure model. Accumulation of contaminants in agricultural meat is described using a simple food chain model, and contaminant concentrations in the four classes of vegetation are determined from estimated equilibrium bioconcentration factors applied either to air or soil. To evaluate the BETR human exposure model in comparison with CalTOX, intake fractions were calculated by both models for individual emissions of 317 chemicals to air in a generic environment calibrated to represent the continental United States. In this comparison exercise the BETR model was simplified to an environmental scenario that can be represented in CalTOX, i.e., the environment, population and food production are described using a single set of homogeneous parameters. Agreement was within a factor of 3 for 250 of the 317 chemicals (79%) and within a factor of 10 for 304 of 317 chemicals (96%), indicating the current model provides results that are consistent with an existing state-of-the-art multi-media human exposure model. Consumption of agricultural foods was the dominant exposure pathway

among the 13 chemicals for which there was greater than a factor of 10 difference between the models, and in all cases the BETR model predicted higher exposure for these chemicals than did CalTOX due to the differences between the models in describing contaminant transfers in the agricultural food chain. Intake fractions calculated by the two models agreed within a factor of 2 for benzene, carbon tetrachloride, 2,3,7,8-tetrachlorodibenzo dioxin and benzo[a]pyrene, which are examined in more detail below.

Spatially explicit input data for North America

The spatially explicit source-to-intake model for North America requires input parameters to describe the environment (e.g. rainfall rate, soil characteristics, vegetation cover), population density and intensity of food production in each region of the model. A database of environmental characteristics and population distribution is already available for the BETR North America regions as part of the contaminant fate model database (Woodfine, et al. 2001). But to extend the spatially explicit assessment to human exposure required compilation of a database of food production activities in each region.

Several assumptions were made to reduce data requirements and simplify the spatially explicit characterization of food production and distribution. We assume that the population of North America only consumes food that is produced on the continent, i.e. there is no consideration of food imports from outside the continent. Additionally, we assume that there are no barriers to transport of food within the continent and that consumers do not show a preference for locally produced foods. This implies that all food produced in North America is combined into a single food basket and that all individuals eat from this food basket. It is emphasized that these assumptions can only be applied in calculations of total population intake, and are not valid for characterizing exposures to specific sub-populations or individuals. However, adopting this approach allows a spatially explicit assessment to be compiled while introducing minimal additional complexity and data requirements. It is consistent with our goal to develop and evaluate models that are iteratively more complex. In this way we assess the

importance of including spatial information in population-based human exposure assessments.

Approximate food production rates for eight food categories in each region have been compiled from available data sources to estimate the relative contribution of production in each BETR region in the exposure assessment (Table 1). These data have been gathered from a variety of sources, including several United States Department of Agriculture (USDA) reports and databases (USDA 2001a, USDA 2001b, USDA 2001c, USDA-ERS 2002a, USDA-ERS 2002b) Canadian Provincial Government reports (ADAFRD 2001) and the United Nations Food and Agriculture Organization (FAO) statistical database (FAO 2002). In general, the quality and quantity of information available to estimate food production values was highest for the US, where detailed and consistent summaries of state-by-state agricultural production are compiled by the USDA, and lowest for Mexico, where the only available data were the FAO estimates of national food production rates. Given the paucity of data for Mexico, estimated national production was arbitrarily distributed equally among the four BETR regions that lie entirely within Mexico. Data for Canada were extracted from summaries prepared by the individual provinces, but data reporting between provinces was not always consistent and some data gaps existed, particularly for the Yukon, Northwest Territories and Nunavut.

For some agricultural products, human consumption is only one of many uses for the product. For example, over a quarter of corn production in the United States goes to non-human consumption applications including fuel production, animal feed, and seed (USDA-ERS 2002a). In addition, a significant fraction is used as raw material to produce highly refined products such as high fructose corn syrup, beverage alcohol and refined sugars, in which it is assumed that contaminants are eliminated during the food processing. Because the individual agricultural products are combined into general food categories, it was necessary to estimate the percentage of production of each product that enters the human food supply from each region before summing with other products. This adjustment corrects the relative production rate between states or provinces that produce products with different proportions of production going to non-human

consumption uses. In the case of corn, for which there are many non-human consumption uses, failure to adjust production values would result in overestimating production of produce in corn-producing regions. Adjustments were also made to correct for the fraction of specific agricultural products that are exported out of the continent (USDA-ERS 2002a).

The accuracy of the production data was evaluated by comparing estimated production rates to estimated population intake rates. Intake rates for individual food categories in the model are based on the USEPA Exposure Factors Handbook (EPA 1997), interpreted to combine individual crops to match the food typed used for production rates. The production rate was higher than the intake rate for leafy vegetables, exposed produce, grains, eggs, dairy and meat, but in all cases within a factor of three. Production rates for root crops and protected produce were similarly within a factor of four of estimated population consumption rates. The discrepancy is reasonable, and can be attributed to losses due to waste in food processing and preparation and spoilage at all stages of distribution and use. Given the uncertainties associated with combining food production estimates into broad categories, and the variability in data quality and availability, the relative production values reported in Table 1 are approximate and should be treated as a “best estimate” of the distribution of regional food production for North America.

Intake Fraction in source-to-intake modeling

Intake fraction is defined as the integrated incremental intake of a pollutant released from a specified source or source class, summed over all exposed individuals and occurring over a given exposure time, per unit of pollutant emitted (Bennett, et al. 2002b). Within a defined region intake by a hypothetical representative individual resulting from spatially averaged exposure concentrations within that region is calculated and extrapolated to the entire population. This “per caput” approach was originally developed by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) to assess population dose commitments from nuclear technologies and radioactive fallout (UNSCEAR 1977). An advantage of a population-based measure is that it accounts for

the number of exposed individuals, which may be large if the chemical travels a long distance in the environment and/or if the population density is high. However, it provides no information on exposure variation among individuals within the population of that region.

When using a multi-media contaminant fate model and a multi-pathway exposure assessment, intakes through multiple routes should be considered. In this paper, we assume that dose response functions relate to applied dose. As a result we sum intake across all exposure routes (inhalation, ingestion, and dermal uptake).

If more than one region is considered in the assessment, as in the 24 regions of the BETR North America model, the population iF can be summed over all regions:

$$iF_C = \sum_{j=1}^{24} iF_j \quad (1)$$

where j represents the region number and iF_C is the intake fraction for the entire continental population. The intake fraction will be different in each region (iF_j) for a given emission scenario, a function of the proximity of the region to emissions, the fate of the chemical in the environment and its pathways for human exposure.

Application to four contaminants in North America

We illustrate the patterns of variability in iF from the BETR North America human exposure model by application to four pollutants. Two are hydrophobic chemicals with low vapor pressures, benzo[a]pyrene (B[a]P) and 2,3,7,8-tetrachlorodibenzo dioxin (TCDD). Two are volatile chemicals, benzene and carbon tetrachloride. Physicochemical properties used in this assessment are shown in Table 2 (CalTOX 2001) along with the Characteristic Travel Distance (CTD) for emissions to air of each chemical, calculated using the TaPL III model (Beyer, et al. 2000). The CTD is the distance pollutants travel in an idealized model environment before their concentrations are reduced by a factor of

2.718, or e, by irreversible deposition and/or degradation. The four chemicals encompass a range of CTDs, with relatively short and long travel distances represented for both hydrophobic and volatile compounds. In North America, emissions of the four chemicals considered here are dominated by releases to air and only this emission scenario is examined here. The emission rate for all chemicals was 1000 kg/year, but this selection was arbitrary since the rate of emissions cancels out in calculating iF.

As a baseline for comparison, the BETR model framework was re-parameterized to represent the entire North American environment as a single region with average characteristics consistent with the spatially resolved 24-region model. This approach is equivalent to that used in existing single region models. The influence of location of contaminant release on iF for the continental population is evaluated by determining the iF value for emissions to air in each region of the spatially explicit model individually. Twenty-four model runs, one for emissions in each region, yield 24 different iF values for exposure to the entire continental population.

RESULTS

Results from the non-spatially differentiated model are presented in Table 3. For both benzene and carbon tetrachloride, inhalation is by far the dominant exposure route for the population. The iF for carbon tetrachloride is an order of magnitude higher than that of benzene due to its longer environmental persistence. The dominant exposure pathway for B[a]P is deposition to vegetation and subsequent consumption by the population, while TCDD exposure is through the agricultural food chain. B[a]P exposure from meat and dairy products is relatively low because it is subject to metabolism by animals in the agricultural food chain, while TCDD is not.

Figure 2 shows iF due to emissions in each region for benzene and carbon tetrachloride, and Figure 3 shows iF for B[a]P and TCDD. For benzene (Figure 2), the calculated iF ranges over two orders of magnitude depending on emission location and the pattern follows population distribution, with the highest iF for release in the most densely

populated regions. Emissions in sparsely populated and remote regions in the North result in markedly lower iF values than emissions in the temperate, more densely populated regions of the continent. Calculated iF values for carbon tetrachloride are less variable with release location because it has a greater CTD, but they still span an order of magnitude with higher values corresponding to release in regions of high population density. Inhalation is the dominant exposure pathway for the continental population for benzene and carbon tetrachloride, accounting for greater than 97% of total exposure for both chemicals for all release locations.

For both B[a]P and TCDD, iF ranges over almost 3 orders of magnitude depending on release location (Figure 3). The spatial pattern of intake fraction follows the pattern of food production. As for the volatile chemicals, the lowest iF is for emissions in the remote regions of the North, where local food production makes a very small contribution to the continental total. The two highest iFs for B[a]P are for emissions in the regions with the highest production of vegetables and grains while the highest iFs for TCDD are for emissions in regions with high proportions of the continental production of meat and dairy products. Location of emission does not significantly affect the dominant exposure pathways for B[a]P, with exposure by consumption of vegetation accounting for greater than 85% of the total in all cases. For TCDD the dominant exposure route is ingestion of agricultural meat, dairy and eggs for emissions in all regions with the exception of Region 1. Region 1 accounts for a high proportion of the continental production of coastal fish, and contributes negligibly to all other food categories. Emissions of TCDD in Region 1 result in fish consumption contributing 86% of the total population exposure, however, population intake fractions for emissions in this region are among the lowest for any release location.

For all four chemicals, the iF calculated by the single region model is very close to the median iF for emissions into the individual regions. This indicates that an appropriately parameterized single-region model will give results that are consistent with the spatially explicit model, but without any information about variability in population iF due the source location.

Regression analysis

Figures 2 and 3 clearly demonstrate that iF calculated for the continental population is a strong function of both the properties of the chemical and where the emission occurs. As discussed above, the variability in iF with release location for each chemical appears to be related to regional differences in population density and intensity of food production. However, environmental variability in characteristics such as atmospheric residence time and surface areas of soil, fresh water and coastal water also contribute to variability in iF because they influence the environmental fate of the chemical.

We applied simple stepwise linear regressions using the coefficient of determination (R^2) to characterize the influence of variability in population density and intensity of food production in the region of release on iF for each chemical. Regressions were initially sought between iF calculated by the model as the dependent variable and the spatially explicit input parameters reported in Table 1 as independent variables. Based on the calculated regression coefficients and the dominant exposure pathways for each chemical, one or two independent variables that accounted for a significant proportion of the variability in iF were selected. Regression equations for the four chemicals are shown in Table 4.

For three of the four chemicals, greater than 75% of the total variation in iF with release location can be accounted for by a linear model with one or two independent variables, as indicated by each R^2 in Table 4. The equations only consider spatial variability in model input parameters between the source regions. Therefore, they are most effective at describing variability in iF for benzene and B[a]P, which have relatively short CTD. These contaminants are not efficiently transported in the environment out of their region of release, but are distributed to the entire continental population in foods produced in the source region.

For the volatile compounds, the magnitude of the R^2 indicates the relative importance of exposure in the region where emissions took place compared to the exposure due to inter-regional transport. Evaluation of volatile chemicals with shorter atmospheric half-lives and characteristic travel distance than benzene, for example 1,3-butadiene, yielded R^2 values greater than 0.9 in regressions against population density in the region of emission. However, even for carbon tetrachloride, which is effectively transported between regions of the model in the atmosphere, the simple linear regression model accounts for a significant amount of the variability due to release location.

DISCUSSION

From a public health perspective it is important to acknowledge that exposures occur through multiple pathways and routes including inhalation, food consumption and dermal contact, and that the relative importance of these pathways and the magnitude of population exposure may be determined by the location of sources. Total population exposure depends on a diverse set of variables that include (1) chemical properties, especially persistence and environmental partitioning, (2) environmental transport and sequestration processes, (3) the proximity of sources to population and (4) the proximity of sources to regions of food production. The interactions between these variables are complex and sometimes non-intuitive. A model such as the one described here, interpreted using intake fraction, is a useful conceptual tool for understanding the relative efficiency with which chemicals migrate through the environment to humans. For the public and policymakers, a spatially explicit assessment is more credible and informative than a single-region assessment that ignores two of the four variables that determine population exposure.

Although uncertainty and variability in chemical properties and environmental conditions are included in uncertainty analyses, the intensity of food production and population density within the study area are often overlooked. These model assumptions can contribute significantly to the overall uncertainty in iF calculated by single region models such as EUSES and CalTOX. Based on the calculations presented here for emissions of

contaminants to air in North America this variability can be 2 – 3 orders of magnitude, depending on the chemical being considered. In general, chemicals that are widely transported in the environment will exhibit lower variability in iF due to location of release because the chemical will not establish strong concentration gradients in the environment. In other words, the common assumption that pollutants are well-mixed within the environment is most applicable when chemicals are transported effectively over the spatial scale of the model region in the assessment.

The magnitude of variability in iF attributable to release location determined here is comparable to the 0.5 – 6 orders of magnitude range of uncertainty in toxicity potentials resulting from uncertainties in fate and transport, exposure, and toxicity using a conventional multimedia model (Huijbregts, et al. 2000b). Therefore, failing to consider spatial variability in assessments of iF may significantly under represent uncertainty in the overall assessment of risk to human health and life-cycle impact assessment.

Moreover, the true spatial variability in population exposure is probably greater than that calculated by the BETR model. For contaminants released to air with inhalation as the dominant exposure pathway, the key parameter determining population exposure is the population density in the area impacted by the emissions. The area affected by emission is a function of the characteristic travel distance of the chemical. In this exercise the model regions of North America are large and are characterized by an average population density. Contaminants with short CTD will have strong gradients of concentration on spatial scales smaller than those described by the model, which will therefore underestimate variability in population exposure by ignoring the overlapping gradients in population density and exposure concentration. In multi-pathway exposure assessments uncertainties are lowest for exposure through inhalation thus increasing the relevance of accurately characterizing the spatial variability of inhalation exposure due to variations in population density. Therefore the application of this model for volatile compounds should be limited to compounds with a spatial range comparable to the size of the region (i.e., not less than approximately 500 km). Ideally, the spatial scale of the region or set of

regions used in a model assessment should correspond to the spatial range of the contaminant.

For compounds where exposure by the ingestion pathway dominates, information on spatial variability in food production rates is essential for compiling an accurate assessment. This information is much less readily available than the population data required for inhalation-dominant chemicals. Additionally, there is much greater uncertainty in the source-to-intake relationship for these chemicals due to the uncertainties associated with contaminant transfer into food. Because of these limitations, we believe the spatial scale of the BETR North America model is appropriate for hydrophobic contaminants released from area sources regardless of their CTD.

In compiling a population-level exposure assessment, the advantages of a spatially explicit model must be weighed against the increased requirements for input parameters and computation over the corresponding single-region model. For screening-level impact assessments at the population level it is likely advantageous to compile the initial assessment using a single region model and then apply a spatially resolved model as required to characterize variability in population exposure as a function of release location. The spatially explicit model can be used to compliment a simpler single-region assessment, and to address specific policy questions. Ultimately, the decision whether or not spatial variability needs to be included in population-level exposure assessments must be made on a case-by-case basis, considering the goals of the assessment and the characteristics of the chemicals under consideration.

It is noteworthy that exposure assessments that do not include spatial resolution are not able to address certain key policy questions. For example, a spatially explicit assessment is necessary to evaluate the relative contribution to total population intake in a remote region due to a small local source versus a larger source in a distant location. The nature and extent of anticipated health effects - whether they are chronic and stochastic or acute and non-stochastic - may also dictate whether a spatially explicit assessment is required. Non-stochastic effects are those for which the severity of the effect increases with dose.

For non-stochastic and acute effects refinement of the initial assessment should focus on more accurately describing the distribution of doses experienced by individuals within the population. Stochastic effects are those for which the likelihood or incidence of a defined effect increases with dose. For stochastic and chronic effects, population intake is the relevant measure of risk and a spatially explicit assessment would be useful to deduce relative population exposures due to sources in different locations.

The regression analysis presented here offers a method to extrapolate from a generic exposure assessment to estimate iF values for a range of population density and food production activities in an assessment region. The regression results indicate that variability in population density and intensity of food production has a greater influence on population exposure than variability in environmental conditions between release locations. In assessments using single-region models, the magnitude of this variability can be estimated from the variability in population density and intensity of food production within the area under consideration.

In summary, a spatially explicit multi-media contaminant fate and human exposure model based on the Berkeley-Trent North American contaminant fate model framework has been developed and applied to four chemicals emitted to the atmosphere in North America. Intake fraction has been employed as a metric to characterize the source to human intake relationship for individual chemicals. Calculated iF can range over almost 3 orders of magnitude depending on release location in the North American continent. The model presented here provides a framework for spatially explicit tracking of contaminant emissions from sources to human intake, and removes some of the conceptual limitations of existing models that describe the environment as a single homogeneous system.

ACKNOWLEDGMENTS

This work was supported by the Natural Sciences and Engineering Research Council of Canada (NSERC) and by the US Environmental Protection Agency National Exposure

Research Laboratory through Interagency Agreement # DW-988-38190-01-0, carried out at Lawrence Berkeley National Laboratory through the US Department of Energy under Contract Grant No. DE-AC03-76SF00098. Dr. Bennett's research is supported by the Kresge Center for Environmental Health, NIEHS ES000002. D.A. Koehler, Y. Nishioka and J. Gouin contributed to this research by compiling data on agricultural production in the U.S. and Canada.

Table 1: Rates and percentages of total continental food production in each region of the BETR North America model in food categories considered in the exposure assessment.

Region	Population (Millions)	Food Production (Millions of kilograms per year and percentage of total continental production)															
		Agricultural Crops								Animal Products							
		Grains ^[A]		Produce ^[B]		Leafy Vegetables ^[C]		Root Crops ^[D]		Total Meat ^[E]		Milk		Eggs		Coastal Fish	
1 Yukon River-Aleutian	0.50	0	0.0%	0	0.0%	0	0.0%	0	0.0%	0	0.0%	6	0.0%	0	0.0%	1931	30.2%
2 Mackenzie River	0.43	754	0.5%	151	0.2%	0	0.0%	0	0.0%	86	0.2%	37	0.0%	2	0.0%	0	0.0%
3 Arctic Archipelago	0.03	0	0.0%	0	0.0%	0	0.0%	0	0.0%	0	0.0%	0	0.0%	0	0.0%	0	0.0%
4 Ungava-Goose Bay	0.05	0	0.0%	0	0.0%	0	0.0%	0	0.0%	0	0.0%	0	0.0%	0	0.0%	0	0.0%
5 Fraser & Skeena Rivers	2.62	237	0.2%	59	0.1%	148	1.6%	215	0.6%	288	0.7%	580	0.7%	45	0.7%	318	5.0%
6 Saskatchewan River	4.82	42724	30.8%	9971	13.4%	12	0.1%	1709	5.1%	3056	7.6%	1824	2.1%	125	1.8%	0	0.0%
7 James Bay Shield	0.42	0	0.0%	0	0.0%	0	0.0%	0	0.0%	0	0.0%	0	0.0%	0	0.0%	0	0.0%
8 Gulf of St Lawrence	10.49	997	0.7%	30	0.0%	0	0.0%	2784	8.4%	1114	2.8%	3008	3.5%	103	1.5%	842	13.2%
9 Columbia River	9.33	8274	6.0%	1797	2.4%	23	0.2%	12126	36.4%	573	1.4%	5478	6.3%	136	1.9%	263	4.1%
10 Missouri & Cheyenne Rivers	12.88	19261	13.9%	3420	4.6%	23	0.2%	1797	5.4%	4850	12.1%	2507	2.9%	296	4.3%	0	0.0%
11 Mississippi-Ozark	16.25	7564	5.5%	8240	11.1%	82	0.9%	1717	5.2%	4346	10.9%	12119	14.0%	536	7.7%	2	0.0%
12 Great Lakes Basin	33.17	4692	3.4%	2679	3.6%	367	3.9%	2226	6.7%	2106	5.3%	12900	14.9%	499	7.2%	55	0.9%
13 Appalachian-Atlantic Coast	56.53	1302	0.9%	1198	1.6%	314	3.4%	1256	3.8%	2618	6.6%	8731	10.1%	627	9.0%	533	8.4%
14 Ohio River-Allegheny	25.52	4287	3.1%	3666	4.9%	100	1.1%	246	0.7%	2674	6.7%	5935	6.8%	868	12.5%	53	0.8%
15 Blue Ridge-Everglades	31.76	988	0.7%	13054	17.5%	403	4.3%	901	2.7%	5351	13.4%	2398	2.8%	733	10.5%	176	2.8%
16 Sierra Nevada-Pacific Coast	32.11	2052	1.5%	5010	6.7%	5470	58.8%	3760	11.3%	518	1.3%	12690	14.6%	405	5.8%	312	4.9%
17 Colorado River	5.28	1481	1.1%	333	0.4%	1564	16.8%	1008	3.0%	592	1.5%	2579	3.0%	46	0.7%	9	0.1%
18 Arkansas River-High Plains	6.80	9254	6.7%	640	0.9%	63	0.7%	577	1.7%	2537	6.3%	1791	2.1%	139	2.0%	6	0.1%
19 Mississippi Delta	8.24	3852	2.8%	1309	1.8%	9	0.1%	177	0.5%	2507	6.3%	1085	1.3%	277	4.0%	509	8.0%
20 Rio Grande	23.88	8555	6.2%	5292	7.1%	303	3.3%	1238	3.7%	2994	7.5%	5250	6.1%	599	8.6%	412	6.5%
21 Baja California	8.60	5625	4.1%	4408	5.9%	156	1.7%	420	1.3%	942	2.4%	2069	2.4%	380	5.5%	240	3.8%
22 Sierra Madre Del Sur	25.87	5600	4.0%	4400	5.9%	90	1.0%	393	1.2%	935	2.3%	1920	2.2%	380	5.5%	240	3.8%
23 Sierra Madre Oriental	25.14	5600	4.0%	4400	5.9%	90	1.0%	393	1.2%	935	2.3%	1920	2.2%	380	5.5%	240	3.8%
24 Yucatan Peninsula	15.09	5600	4.0%	4400	5.9%	90	1.0%	393	1.2%	935	2.3%	1920	2.2%	380	5.5%	240	3.8%
Total	355.80	138701	100%	74458	100%	9306	100.0%	33336	100.0%	39957	100.0%	86748	100.0%	6957	100.0%	6383	100.0%

[A] Barley, corn processed as grain, oats, millet, rice, rye, sorghum and wheat

[B] Beans, soybeans, peanuts, pumpkin, sweet corn, fresh beans, melons, nuts, oranges, grapefruit and other citrus, berries, apples, pears, cherries, grapes, peaches, apricots, plums and prunes, strawberries, cucumber, squash, tomatoes and peppers

[C] Lettuce, cabbage, cauliflower, broccoli, spinach and other greens, celery

[D] Potatoes, sweet potatoes, onions, carrots and garlic

[E] Beef, chicken, turkey, ducks, hogs and sheep

Table 2: Physicochemical properties and Characteristic Travel Distance (CTD) of selected chemicals.

	Benzene	Carbon Tetrachloride	Benzo[a]pyrene	2,3,7,8- Tetrachlorodibenzo dioxin
Molar Mass (g/mol)	78.11	153.8	252.3	321.98
Log Kow	2.18	2.72	6.34	6.66
Log Koa	2.81	2.69	10.8	9.67
Log Kaw	-0.63	0.03	-4.46	-3.01
Media-specific degradation half life (hours)				
Air	142	86700	1.5	720
Vegetation	4560	86700	5490	52400
Fresh water	268	6480	56.1	10300
Coastal water	238	6480	56.1	10300
Soil	4560	4720	5490	160000
Sediment	536	4460	28100	49400
CTD (km)	2,944	1,720,543	31	9,627

Table 3: Intake fraction (iF) and exposure pathways calculated using a single-region North American model with characteristics consistent with BETR North America.

	Population iF x 10 ⁶	Percentage of Total Population Exposure by Pathway					
		Aquatic Foods	Vegetation and soil	Meat, Dairy and Eggs	Inhalation	Drinking water	Dermal
Benzene	0.80	0.01	0.08	0.47	99.37	0.07	0.00
Carbon tetrachloride	3.1	0.02	0.03	0.38	99.54	0.04	0.00
B[a]P	92	0.01	96.84	2.93	0.21	0.00	0.00
TCDD	1139	0.95	17.69	81.28	0.04	0.00	0.00

Table 4: Regression analysis of variability in $iF \times 10^6$ with release location in the BETR North America model.

Regression Model	R^2
$iF_{\text{Benzene}} = 0.410 + 0.0378 * P_{\text{dens}}$	0.90
$iF_{\text{Carbon tetrachloride}} = 2.82 + 0.054 * P_{\text{dens}}$	0.45
$iF_{\text{B[a]P}} = -8.87 + 0.074 * \text{Leafy} + 0.012 * \text{Grains}$	0.92
$iF_{\text{TCDD}} = 606 + 0.24 * \text{Meat} + 0.10 * \text{Milk}$	0.75
<p>P_{dens} = Population density in region of emission (persons / km^2)</p> <p>Leafy = Intensity of production of leafy vegetables in region of emission ($\text{kg} / \text{km}^2 / \text{year}$)</p> <p>Grains = Intensity of grain production in region of emission ($\text{kg} / \text{km}^2 / \text{year}$)</p> <p>Meat = Intensity of meat production in region of emission ($\text{kg} / \text{km}^2 / \text{year}$)</p> <p>Milk = Intensity of milk production in region of emission ($\text{kg} / \text{km}^2 / \text{year}$)</p>	

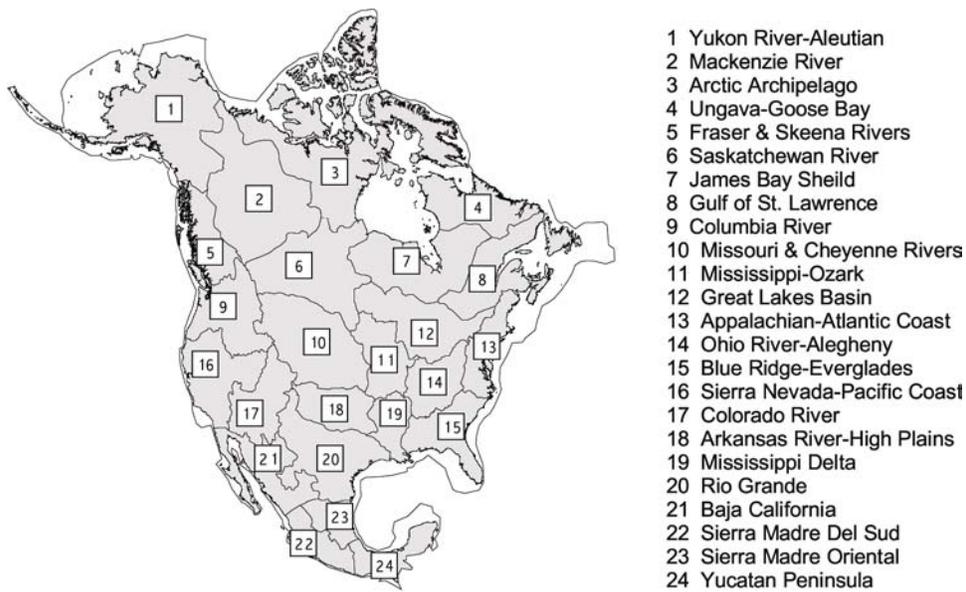


Figure 1: Regional segmentation of the BETR North America model.

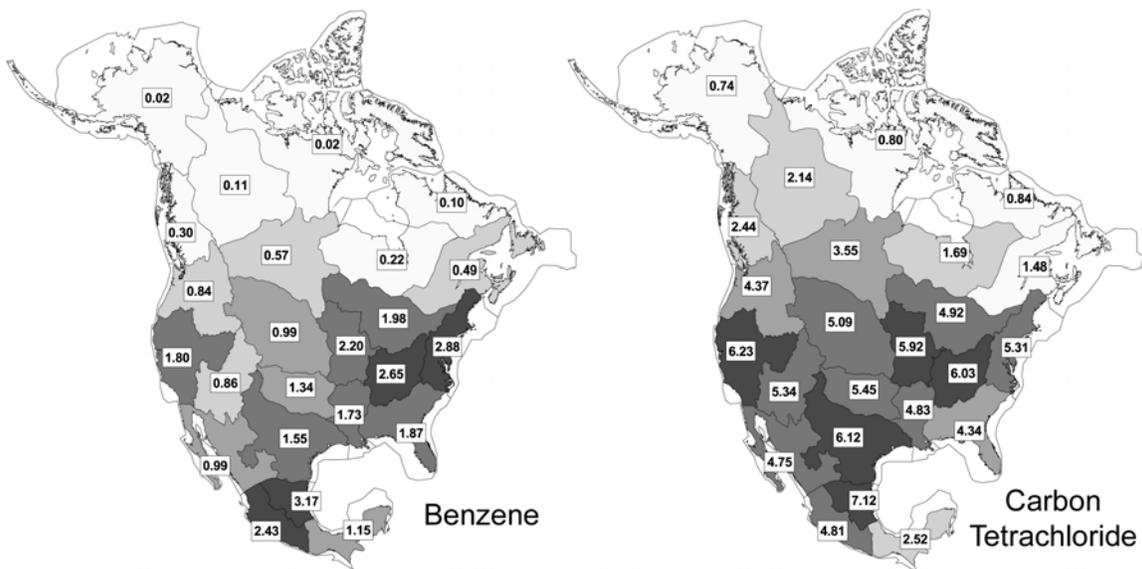


Figure 2: Continental population intake fraction (times 10⁶) for emissions of benzene and carbon tetrachloride to air in individual regions of the BETR North America model. Darker shading represents higher population intake fraction for emission into that region.

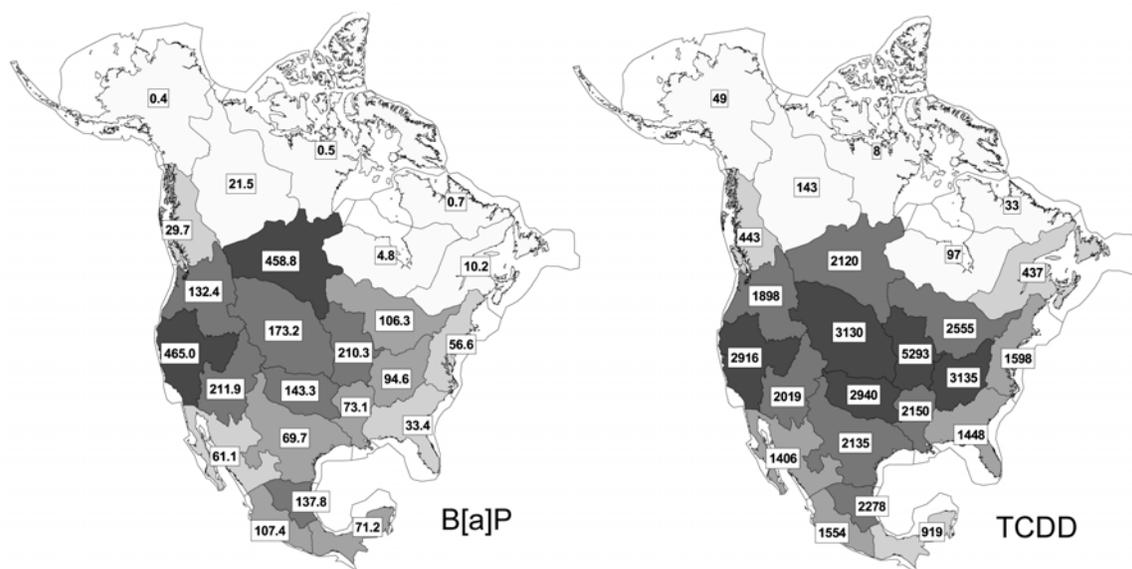


Figure 3: Continental population intake fraction (times 10⁶) for emissions of B[a]P and TCDD to air in individual regions of the BETR North America model. Darker shading represents higher population intake fraction for emission into that region.

References

- ADAFRD. 2001. *Government of Alberta Department of Agriculture, Food and Rural Development - Agriculture Statistics Yearbook*, Available online, <http://www.agric.gov.ab.ca/economic/yearbook/index.html>.
- Bennett D., M. Margni, T. McKone and O. Jolliet. 2002a. Intake fraction for multimedia pollutants: A tool for life cycle analysis and comparative risk assessment. *Risk Analysis* 22: 905.
- Bennett D. H., W. E. Kastenbergh and T. E. McKone. 1998. A multimedia, multiple pathway risk assessment of atrazine: the impact of age differentiated exposure including joint uncertainty and variability. *Reliability Engineering & System Safety* 63: 185-198.
- Bennett D. H., T. E. McKone, J. S. Evans, W. W. Nazaroff, M. D. Margni, O. Jolliet and K. R. Smith. 2002b. Defining intake fraction. *Environmental Science & Technology* 36: 206A-211A.
- Beyer A., D. Mackay, M. Matthies, F. Wania and E. Webster. 2000. Assessing long-range transport potential of persistent organic pollutants. *Environmental Science & Technology* 34: 699-703.
- Brandes L. J., H. den Hollander and D. van de Meent. 1996. *SimpleBox 2.0: A nested multimedia fate model for evaluating the environmental fate of chemicals*. 719101029, RIVM, The Netherlands.
- CalTOX. 2001. *CalTOX 4.0: A multimedia contaminant fate and multipathway human exposure model*, Lawrence Berkeley National Laboratory Exposure and Risk Assessment Group, <http://eetd.lbl.gov/ied/ERA/>.
- EPA. 1997. *Exposure Factors Handbook*. <http://www.epa.gov/ncea/pdfs/efh/front.pdf>, EPA National Center for Environmental Assessment Office of Research and Development, Washington DC.
- FAO. 2002. *United Nations Food and Agriculture Organization statistical database*, Available online, <http://apps.fao.org/>.

- Hertwich E. G., S. F. Mateles, W. S. Pease and T. E. McKone. 2001. Human toxicity potentials for life-cycle assessment and toxics release inventory risk screening. *Environmental Toxicology and Chemistry* 20: 928-939.
- Huijbregts M. A. J., U. Thissen, J. B. Guinee, T. Jager, D. Kalf, D. van de Meent, A. M. J. Ragas, A. W. Sleeswijk and L. Reijnders. 2000a. Priority assessment of toxic substances in life cycle assessment. Part 1: Calculation of toxicity potentials for 181 substances with the nested multi-media fate, exposure and effects model USES-LCA. *Chemosphere* 41: 541-573.
- Huijbregts M. A. J., U. Thissen, T. Jager, D. van de Meent and A. M. J. Ragas. 2000b. Priority assessment of toxic substances in life cycle assessment. Part 2: assessing parameter uncertainty and human variability in the calculation of toxicity potentials. *Chemosphere* 41: 575-588.
- MacLeod M., D. G. Woodfine, D. Mackay, T. McKone, D. Bennett and R. Maddalena. 2001. BETR North America: A regionally segmented multimedia contaminant fate model for North America. *Environmental Science and Pollution Research* 8: 156-163.
- McKone T. E. 1993. *CalTOX, a multimedia total exposure model for hazardous waste sites. Part 1: Executive summary*. UCRL-CR-111456PTI, prepared for the Department of Toxic Substances Control, California Environmental Protection Agency, Lawrence Livermore National Laboratory, Livermore, CA.
- UNSCEAR. 1977. *United Nations Scientific Committee on the Effects of Atomic Radiation 1977 Report to the General Assembly*, Available by request, <http://www.unscear.org/>.
- USDA. 2001a. *United States Department of Agriculture National Agricultural Statistics Service Index of Estimates*, Available online, <http://www.usda.gov/nass/pubs/estindx.htm>.
- USDA. 2001b. *United States Department of Agriculture National Agricultural Statistics Service Index of Estimates, Noncitrus Fruits and Nuts 2001 Preliminary Summary*, Available online, <http://www.usda.gov/nass/pubs/estindx.htm>.

- USDA. 2001c. *United States Department of Agriculture National Agricultural Statistics Service Index of Estimates, Vegetables 2001 Summary*, Available online, <http://www.usda.gov/nass/pubs/estindx.htm>.
- USDA-ERS. 2002a. *United States Department of Agriculture Economic Research Service 2002 Feed Outlook*, Available online, <http://www.ers.usda.gov/publications/so/view.asp>.
- USDA-ERS. 2002b. *United States Department of Agriculture Economic Research Service, Fruit and Tree Nuts, Situation and Outlook Yearbook (October 2001)*, Available online, <http://www.ers.usda.gov/publications/so/view.asp>.
- Woodfine D. G., M. MacLeod, D. Mackay and J. R. Brimacombe. 2001. Development of continental scale multimedia contaminant fate models: Integrating GIS. *Environmental Science and Pollution Research* 8: 164-172.